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ABSTRACT

In 1975 the W. J. Barrow Research Laboratory, Inc., under the sponsorship of the Council on Library Resources, began a series of studies on the chemical and physical properties of book papers between 1507 and 1949. Their testing and review of the literature were concerned with book paper durability and permanence, reasons for decline in durability, methods for predicting paper durability, specifications for durable paper, processing changes needed to produce such paper, and work necessary to preserve weak papers. A history of paper production along with chemical tests showed the reasons for decreased durability; and the feasibility of producing durable paper was demonstrated. Restoring deteriorating books has also proved possible, but at a high cost. A bibliography, testing details, and the table have been included. (LS)

Permanence/Durability of the Book – VII

Physical and Chemical Properties of Book Papers, 1507-1949

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Introduction

Many record custodians have observed that old book papers are often in a good state of preservation. In contrast, newer book papers tend to be weak and brittle. To our knowledge, previous to this study, no one had determined the chemical and physical reasons for this steady decline in paper permanence and durability. Until this information was gathered, it would have been difficult to predict the useful life of paper and to produce at affordable cost and on a commercial scale a paper at least as permanent and durable as some of the fine book papers made by hand centuries ago.

In 1957 William J. Barrow initiated a series of studies, sponsored by the Council on Library Resources, and completed after his death by members of the staff of the W. J. Barrow Research Laboratory, Inc., of Richmond, Virginia. This report presents the findings of these studies, the object of which was to determine from chemical and physical testing, and from a review of the literature:

1. The permanence and durability of book papers produced during the period from 1507 to 1949
2. The reasons for a decline in permanence and durability during this time period
3. An accelerated ageing method for predicting permanence and durability of book papers
4. Specifications for permanent/durable book paper
5. The changes in papermaking processes that would be necessary to produce commercially acceptable permanent/durable paper
6. The further work necessary to conserve weak papers

Work Plan. Part of the technical work described herein has been previously reported.[2,3,4,22,23] Additional tests have now been made to give a deeper insight into the reasons for the gradual decline in paper quality and to suggest how permanent/durable paper can be made. In 1971 a thorough and scholarly report on the history of permanent/durable bookpaper was published by Verner W. Clapp.[7] It is suggested that the reader refer to this publication for background material. Expanded test results from *Strength and Other Characteristics of Book Papers 1800-1899*,[3] by the W. J. Barrow Research Laboratory, and *Deterioration of Book Stock—Causes and*

Remedies[22] and *The Manufacture and Testing of Durable Book Papers*,[23] both the results of studies conducted by W. J. Barrow and published by the Virginia State Library, are included in revised tables in Appendix E, since complete data are necessary to substantiate this report's recommendations, observations, and conclusions.

Definitions and brief descriptions of tests used in this study are given in the next section. Test specimens were taken from 1,470 books published between 1507 and 1949. Each of these books is identified in book lists, which appear as Appendix D.

Technical Descriptions

Definitions. In 1933 the Committee on Permanence and Durability of the Technical Association of the Pulp and Paper Industry (TAPPI) defined *permanence* as "the degree to which paper resists chemical action which may result from impurities in the paper itself or agents from the surrounding air" and *durability* as "the degree to which a paper retains its original qualities under continual usage."[15] The record custodian is interested in both factors, which are commonly referred to jointly as permanence/durability.

Physical and Chemical Tests Used.

1. *Folding endurance.* Performed by using the Massachusetts Institute of Technology Fold Endurance Tester, this test simulates the stresses placed on a book leaf in flexing it to and fro by page turning.[16] It is perhaps the most important physical test in evaluating permanence and durability of book papers.
2. *Tear resistance.* This test carried out by the Elmendorf Tear Resistance Tester, simulates the tear stresses imposed on the pages of a book when they are turned.[18]
3. *Alum, rosin, and groundwood content.* The presence or absence of these three substances, all components or additives that can be detrimental to the permanence and durability of paper, can be indicated by the use of spot tests described by Barrow.[2] Alum and rosin are the components of the most commonly used sizing material. Also, alum is often used alone to simplify the papermaking process. Alum, and to a lesser degree, rosin, have an acid reaction that weakens paper fibers. In addition, the presence, even in small percentages, of groundwood can adversely affect the permanence of paper. Significant percentages can lower the durability.
4. *Metal carbonates.* These mildly alkaline substances tend to counteract any acid that paper may originally contain or later acquire from its environment. The hard water commonly used in making older papers contained dissolved limestone (a form of calcium carbonate). Also, rag-based paper was often made from rags fermented with milk, which contains calcium. Calcium carbonate is deliberately added to modern permanent/durable papers.

   To determine the presence or absence of carbonates, the test

specimens were first immersed in distilled water to expel trapped air and then immersed in about 4% aqueous hydrochloric acid solution. If an evolution of bubbles (carbon dioxide from the carbonate) from the surface of the paper was observed upon immersion in the acid solution, the test was considered positive for metal carbonates.

5. *pH.* Acidity or alkalinity is expressed in pH units. pH 14 is highly alkaline, pH 7 is neutral, and pH 1 is highly acidic. The pH scale is logarithmic; a difference of one pH unit represents a ten-fold difference in acidity or alkalinity. The pH of the paper samples was determined by the TAPPI cold extraction method, using a Beckman Zeromatic pH Meter.[17]
6. *Fiber analysis.* Standard microscopic fiber analysis techniques were used to determine either the type of fiber (hardwood, softwood, rag, and so on), or the length of the fibers, or both, in the paper being examined.

The details of the physical and chemical test methods, and a list of the abbreviations and terms used in the tables, can be found in Appendix A.

Papermaking before 1507

This brief description of ancient and pre-1507 papermaking is intended to introduce the reader to some of the techniques inherited over the centuries by those whose papermaking is the principal subject of this study.

In 105 A.D. the Chinese were the first to produce true paper. which they did by macerating vegetable fibers such as the bark of trees and the filaments of plants and grasses including mulberry and hemp to a pulp in water with a mortar and pestle. Originally, the pulp was probably dipped from the vat, poured into the mold, and allowed to drain and dry in the sun. But within a short time, improved methods were developed and adopted.[9] After maceration the fibers were lifted from the slurry with a screen or mold to form a matted residue—paper—of improved quality and uniformity. Much later, the process of drying and forming the paper by transfer from the mold to felt—couching—was invented.

Sizing, a means of decreasing or eliminating feathering of ink when applied to paper to allow for clear, sharp printing or writing, was developed in China by 700 A.D. Gypsum was used first, followed by starch or rice flour, and then lichen glue. The early Chineses paper was burnished by hand rubbing with agate or some other type of smooth, hard stone.[9]

The art of papermaking traveled to Korea and Japan by 600 A.D., then to Samarkand in 713 A.D. via Chinese prisoners of war. From Samarkand, it entered the Arab world—Baghdad in 793, Egypt in 900, and Morocco in 1100.

The Arabs modified the Chinese technique by fermenting heaps of wet rags, then boiling the fermented mass in wood ashes. The dirt and alkaline residue remaining after this process was at least partially removed by suspending bags of fiber into a flowing stream. In India, rags were fermented with lime for several days.[9] In all probability, the wood ash and lime treatments left an alkaline residue, which would have contributed to the paper's permanence and durability.

The Moors invaded Spain in 711 A.D. and introduced papermaking to that country shortly thereafter. Hundreds of years after its origin, papermaking had finally entered Europe. The Spainards modified earlier fermentation methods and heaped moist rags for some two months. This method often rotted one-third of the cloth beyond use, although the remaining fiber was

tender and easily beaten.[9] The Spainards also adopted the water-driven stamper from the Chinese and Arabs (some maintain that the Spanish invented the stamper). This device was used to macerate the raw materials to a fibrillated pulp. The fiber length was apparently not altered significantly by the stamper.[14] Spanish modifications of Arabian and Chinese techniques included the use of metal instead of vegetable filament molds and the introduction of felt (a matted substance composed of hair and wool) to couch the paper sheets.

From Spain, the knowledge of papermaking skills traveled to Italy. Italian paper was manufactured at the Fabriano mills in 1276. There the first known European watermark appeared in 1282.

Of importance in this chronology is the first use of gelatin size in 1337. Animal gelatin size (the first type of gelatin size) was produced from horns, hooves, and hides of animals and was applied to the formed paper sheet by soaking it in a hot solution of the gelatin.

Originally, sizing was designed to be used mainly on papers that were to be written upon by quill pens. Some old printing inks were evidently of such composition that very little sizing was needed. The paper in many of the earliest books contained no sizing at all.[8]

From Italy, papermaking spread to France in 1348, Germany in 1390, Flanders in 1405, Poland in 1491, and England in 1495.

Papermaking received impetus from the popularization of printing. The advent of large-scale printing and the consequent increased use of paper, however, did not develop until completion of Johann Gutenberg's Bible in 1455. From that date, printing and the need for paper increased at a rapid rate. By 1487, some type of printing was being carried out in all European countries.[8] Although permanent/durable paper had been made for centuries, the spread of papermaking to Europe from the Orient was retarded by reluctance to accept its presumed lack of permanence and durability as compared to parchment. In 1494, Trithemius, Abbott of Sponheim, asked: "If writing is inscribed on parchment it will last for a thousand years. But if on paper, how long will it last? Two hundred years would be a lot."[21]

In some respects, the abbott was a prophet; once the art of papermaking gained acceptance (hastened by the great

revolution in learning through book printing), only sporadic attention was given to the quality of the paper on which lasting words were to be printed.

An Englishman, John Murray, decried in 1829 the fact that a Bible, only thirteen years old, was already "crumbling literally into dust."[13] Since that time, many individuals and organizations have studied the problem of paper permanence, with incomplete or erroneous conclusions.[7]

European Papermaking from 1507 to 1699

Major developments of the 1600s involved beating, pressing, and sizing. In the early 1600s stampers became more sophisticated. Instead of wooden mallets, three types of stampers were used to perform operations on the pulp: (1) pestles shod with iron teeth or spikes for fraying, (2) stampers having finer steel teeth to achieve further maceration, and (3) wooden stampers without teeth to complete the beating process. Washing between stages removed impurities.[9]

Stampers were inefficient and soon were replaced by a newer invention called the Hollander beater. The exact date of this invention is somewhat nebulous, but Henk Voorn asserts that the device was invented between 1630 and 1655 and was modified in 1673.[24]

The original Hollander beater was a tub or vat in which the rag-and-water slurry was circulated and beaten by the mechanical interaction of the beater roll (fitted with thirty iron knives) and the bedplate. According to Voorn, the modifications made in 1673 entailed the replacement of the iron knives and bedplates with ones made of a mixture of brass and silver. It was probably this modification that allowed the Dutch papermakers to manufacture their famous "white" papers, which unlike other contemporary papers, were not stained with the color of iron rust as a result of corrosion of the hardware.

It is not clear when the press (used to remove water from the paper sheet) was invented, but it is known that such a device was in use in the early 1500s. By 1540, the glazing hammer replaced hand burnishing. The hammer, as wide as the paper sheet and operated by a water wheel, left a smoother and more uniform surface on the formed sheet.

Although records indicate that the use of gelatin sizing began in 1337, there is no mention in the available literature of the use of alum with gelatin until the 16th century. In our tests, alum was found in twelve of the forty-five test books from the 16th century.

The first published record of the use of alum alone occurs in a papermaking handbook published in 1634. In this reference, Sung Yinghsing describes a Chinese method of soaking bark papers in alum liquor.[6] Further reference to the use of alum was found in the *Diary of John Evelyn* published in 1678.[8]

Alum was added to gelatin size for three apparent reasons: (1)

to stabilize the viscosity of the size at various concentrations and temperatures, (2) to act as a preservative by inhibiting the formation and growth of bacteria and molds, which putrify the gelatin, and (3) to increase the gelatin's resistance to ink penetration.[12]

Data, Analysis, and Discussion: 1507-1699. Our findings concerning paper manufactured in the period from 1507 to 1699 will be discussed in this section, and the results of changes in method of manufacture that started about the middle of the 17th century will be brought out. Test results showed that the physical and chemical properties of the papers produced during the period from 1507 to about 1610 were reasonably uniform. The remainder of the 17th century was the beginning of a period of change, to the detriment of paper.

Noteworthy test results can be summarized as follows:

1. Especially considering the age of these papers, their fold and tear resistances were surprisingly high, especially as compared to the same characteristics of paper produced in subsequent centuries.
2. The basis weight of paper produced during this period was surprisingly uniform for a hand manufacturing operation.
3. The pH values of the papers remained satisfactory during the 16th century, but began to decrease in the 17th century, especially in papers produced beginning about 1650. This result seems consistent with the increase in positive tests for alum and the negative tests for carbonate in papers made since that date.
4. The gradual decrease in fold resistance and, to a lesser degree, in tear resistance, beginning with or shortly after 1650, can possibly be attributed to the increasing use of the Hollander beater and the "improved" stampers. This supposition is not clearly substantiated by tests of fiber length, but the Hollander and the stampers may have resulted in some other detrimental change in fiber configuration or chemical components of the paper, or may have caused modifications in further steps of the manufacturing process. In any event, changes in paper strength began at about the time the Hollander and the new stampers were introduced (as indicated by the dotted vertical line at about 1670 in figures 2 and 3). Whether or not the simultaneity of these developments was more than coincidence will probably never be known definitely.

Alum and Acidity. The increasing acidity of the 16th-and 17th-century papers became most pronounced early in the 17th century. For papers made in the decade from 1600 to 1609, the median pH value was 6.8 (almost neutral) when tested in 1967. For papers manufactured during the next decade, 1610-1619, the median pH had dropped to 5.4 (somewhat acid). This trend continued almost without exception for the remainder of the century. (The fact that so few exceptions were found may be attributed to the relatively small number of test books available.) This trend toward lower pH values is substantiated by examination of table 2 of Appendix B.

The chemical reasons for this increase in acidity are also obvious from the data in this table 2 and in figure 1 of Appendix C. For example, 80 percent of the papers tested from the decade 1600-1609 contained no alum. Only 20 percent of the tested papers from the next decade were alum-free. This trend persisted during the century, and an overwhelming percentage of the papers made from 1660 to 1699 contained alum. Since acidity is probably the greatest cause of weakness in paper and alum is a major source of acidity, the correlation with decreasing fold and tear resistance is predictable.

As the 17th century progressed, papermakers apparently found that the use of alum was a convenient crutch in the manufacture of paper—a trend which has continued in many cases to the present day.[14] The addition of alum in papermaking tends to encourage the attraction of fibers to each other, thus bringing about the formation of a sheet which otherwise would be difficult or impossible to manufacture because of the nature of the pulp. During this century, alum was used also to "set" the gelatin size, the use of which was becoming increasingly popular. This early use of alum began a trend that was to decrease the permanence and durability of paper.

Carbonate. If we assume that 17th-century papermakers were unaware of the detrimental effect of alum, we must also assume that for some reason, probably related to the increasing demand for paper and consequent shortage of rags, the beneficial effects on paper of calcium and magnesium carbonates content were unknown or ignored. For the 16th century, when the use of alum was much less frequent than in the 17th century, 24 percent of the papers tested contained carbonates. Most of the carbonates in these papers probably came from the lime used to ferment

rags and, to a lesser extent, from the water used in papermaking. During the 17th century, and especially beginning with 1650, the percentage of papers containing these carbonates decreased markedly. This fact is significant because in the absence of carbonate, which is an alkaline material, there was no chemical counterbalance to the presence of acid. Only 7 percent of the papers tested from the 17th century contained carbonates. The negative effect of alum was not balanced by the positive effect of carbonates. Consequently the acidity increased, and the paper became weaker. The weakness of acid papers may not have been apparent to a papermaker of the 17th century, but after three centuries the increase in acidity and associated decrease in strength have become serious. (See tables 2, 3, 4, and 5 of Appendix B, and figure 1 of Appendix C.)

There is some indication that after the invention of the Hollander beater, fermentation of rags ceased at some mills. It seems logical also that the soaking of rags in milk (which contains carbonate-producing constituents) would be used less frequently because of cost and the need to save manufacturing time as the demand for paper increased. The Hollander apparently produced useable fibers from rags without the time-consuming treatment of the rags employed previously. Here again, an improvement in technology was not accompanied by an understanding of the negative side effects on the product.

European and American Papermaking from 1700 to 1799

The 18th century witnessed an escalating need for paper and an increasing change in methods of papermaking, which resulted in a decrease in paper quality. It seems reasonable to assume that the pressure for greater production and a lack of understanding of the consequences of the technological changes played an important role in quality reduction. Papermaking was still an art, and sharing of technical information among papermakers, especially with operators of the new paper mills, was probably essentially nonexistent.

In the first half of the 18th century, the Hollander beater was introduced throughout Europe. It was brought to the United States about 1780. For a time, stampers and Hollander beaters were used simultaneously—stampers for breaking down the rags initially and Hollanders for completing the beating operation. However, the beater had a much higher production than the stampers, and by the end of the century, Hollanders had all but replaced stampers. By about 1740, the shortening effect on fiber length, though slight, began to become apparent. Other effects on the fiber structure, such as change in fibrillation, may have resulted but such effects were not substantiated in our studies.

The method of sizing with gelatin and alum apparently also changed during this century, but authorities disagree as to when this change took place. The old method comprised dipping the finished paper into a vat of gelatin. The new method, called engine sizing, involved incorporation of gelatin and alum into the slurry of fibers and water before the sheet was formed.

Shortage of Rags. The increasing demand for paper resulted in a great scarcity of useable rags as raw material. However, two technological changes occurred during the 18th century to alleviate this problem.

The first was the use of a new method for bleaching colored rags so they might be used in production of light-colored paper. The old method of bleaching with sour milk was too mild to make the use of colored rags feasible. Next, bleaching with sulfuric acid was attempted, with undetermined but predictably detrimental results on paper quality. Then, soon after the discovery of chlorine by Scheele in 1774, this powerful gas was used as a bleaching agent. According to John Murray, chlorine

was "generally clumsily or incautiously or unwarily employed" in such ways that the acidic and oxidative conditions accompanying its use severely weakened the paper sheet.[13] Nevertheless, chlorine bleaching was practiced in England by the end of the century and was brought to the United States shortly thereafter by Joshua Gilpin and used in his Brandywine, Pennsylvania, paper mill. Technological improvements in the use of chlorine and its derivatives for bleaching paper stock have continued to the present day.

The second attempt to overcome the shortage of useable rags was a search for other fibers. This activity increased during the last thirty years of the century. Fibers which were examined included linen, cotton, wood, and straw.[7] The cotton gin was invented in 1793 and the very rapid increase in production of cotton caused the deferral for some decades of research on other fibers.

The 18th century marked the beginning of a period of technological innovation, which, unfortunately, was conducted in an atmosphere of secrecy and without knowledge of the effects of the technological changes that took place. We do not know what methods of beating, stamping, sizing, or bleaching were used on any of the 250 18th-century books in our test sample. However, whatever processing combinations were used, the quality of paper was severely reduced. Unfortunately, the worst was yet to come.

Data, Analysis, and Discussion: 1700-1799. The sacrifice of paper quality for quantity is most noticeable in our results on fold and tear resistances and on pH. This trend is shown dramatically in figures 2 and 3 for fold and tear resistances and in figure 4 for pH change. The decrease in fold and tear resistances was so great that the data had to be presented on a logarithmic scale in figures 2 and 3. The change in pH, already a logarithmic function, is presented in a linear scale in figure 4. The specific median values, by decade, within the century are shown in table 2 of Appendix B. In our experiments the median number of folds before breaking for papers produced in the first decade was 505, in the last decade, 72; the median force required to tear a page dropped from 41.6 to 34.2 grams over a similar time period; and the pH dropped from 5.3 to 4.5, almost a ten-fold increase in acidity. Because of the dates of the particular book samples used, there are intermediate values during

the century, but the downward trend in quality is unmistakeable.

Yet, fortunately for the librarian, archivist, and conservator, many 18th-century papers are still in a relatively good state of preservation as measured by their fold and tear resistances. However, their acidity is quite high, especially in the case of papers produced in the latter half of the century, because of the increasing use of alum and the lower carbonate content. The test findings imply that if these papers are to be preserved for posterity, they should be deacidified in the not too distant future. If they are not deacidified, they will approach a level of strength so low as to require restoration by lamination or other means.

Of the 250 books tested from the 18th century, 213 (85 percent) contained alum. Only 22 (8.8 percent) contained carbonate, and all but one of these were books published between 1700 and 1750. Similarly, all but 6 of the books that gave negative tests for alum were published in the first half of the century. On the basis of our work, therefore, 18th-century books published after 1750 should give the greatest cause for concern. An average pH of about 4.5 for the papers made from 1750 to 1799 leaves us with a great sense of insecurity.

Papermaking from 1800 to 1899: Rapid Technological Change

If the study of 18th-century papers left us with a feeling of uneasiness, the results of the tests of 19th-century papers pointed toward impending catastrophe. Consumer demand placed intense pressure on paper manufacturers to increase production during the 19th-century. Inventions followed, the effects of which on quality were in too many cases unknown or ignored.

By the 19th century, much more rapid methods of producing paper were required. The Fourdrinier and cylinder papermaking machines were invented, improved, and patented between 1800 and 1810. By 1823, it was reported that there were thirty-five such machines in England and others in France and the United States. Their greatly increased rate of production forced the invention of heated drying cylinders since the old "loft" method of hanging paper in the air to dry had become impractical.

Increasing demand for paper at the lowest feasible cost also led to the invention of alum-rosin sizing to replace gelatin sizing. This invention, made by Illig early in the 1800s came into common use beginning about the fourth decade of the century.[10] This size, which is still used extensively by papermakers, is prepared by dissolving rosin in an alkali and, after mixing the solution with the paper fibers, precipitating it by the addition of alum, thus forming an aluminum rosinate, which is an effective sizing material. The sized paper tends to become somewhat brittle with age, probably due to the oxidation of the rosin by air. Because of the low cost and simplicity of this method of sizing, it rapidly replaced gelatin, and all but about 3 percent of the papers tested from the last three decades of the 19th-century contained rosin.

The addition of more alum to precipitate the rosin sizing caused serious problems for the permanence and durability of the papers because of the further increase in acidity. The average pH of the papers produced in the last three decades of the century was low; no carbonate was found in papers of this period to compensate for the acidity; and the fold and tear resistances were much lower than those for earlier papers. There were, of course, other reasons for the poor quality of the paper.

One other possible factor was the Jordan refiner,[14] invented in 1858, which came into increasing use as a supplement to, and

later as a replacement for, the Hollander beater. The Jordan has a conical steel-bladed core revolving within a hollow steel shell. Inside the shell are alternate steel knives and fillers. Using the Jordan, the "stuff" (slurry) could be transferred directly to the paper machine. The Jordan may have been partly responsible for producing a shorter fiber length, which led to weaker paper.

Wood Fibers. Much more detrimental to quality, however, was the use of wood as a source of paper fibers. Wood was cheap and plentiful, and in the papers we tested which were made beginning about the middle of the 19th-century, wood fibers began to appear. In tables 27-31 of Appendix E, the percentages of various types of fibers used to make paper from 1850 through 1899 are shown. By the end of the century, wood fibers were present in a very high percentage of the papers tested. The average fiber length was notably less, and the fold and tear resistances had decreased markedly.

There is nothing inherently wrong with cellulose derived from wood as a papermaking fiber, provided that the wood cellulose is freed of impurities and the fiber length is adequate. Unfortunately, many decades were required to learn how to process wood to give satisfactory paper fibers.

In the 1840s when the use of wood for making paper began, the wood was ground against a revolving wet grindstone to separate fibers. Later a chip grinder was invented. The fibers were not purified, and contained approximately equal amounts of cellulose and noncellulose materials, mostly lignin. The noncellulose components did not have good fiber-bonding properties, were somewhat acidic, were not white, became darker when aged (especially when exposed to light), and contributed no strength. The first groundwood paper was probably much less acceptable than today's newsprint, which contains a proportion of chemical pulp fibers. Consumers did not immediately accept this type of paper, but its low cost and good printability gradually won out.

Following the initial appearance of groundwood papers, improvements in technology gradually permitted the production of wood fibers of increasing cellulose content and lighter color by removing lignin and other impurities. The soda process, the sulfite process, and the sulfate process were developed at the end of the 19th-century.[1] These processes and their modifications had as their objective the solubilization of noncellulose

ingredients with chemicals, usually in water solution at elevated temperatures and pressures, leaving fibers of greatly increased cellulose content. Modifications and improvements in bleaching with chlorine and its derivatives allowed production of lighter-colored and stronger papers.

All of these technological changes took place in a century when science was just beginning to emerge as a potent and controlling force in understanding and changing the properties of matter, and when industry was more enamored with quantity and profit than with quality of product. From the standpoint of the librarian, archivist, and conservator, as well as of the public, the effect of this combination of factors was sad indeed.

Data, Analysis, and Discussion: 1800-1899. The determination of groundwood content in 19th-century papers was made by fiber analysis, not by the spot test used with 20th-century papers. The trend toward increased use of groundwood and rosin is obvious from our test results.

Summary data giving the results of our tests can be found in Appendix B (tables 2, 3, 4, and 5) and Appendix C (figures 1 through 5). In Appendix E, tables 22-31 give the individual results for each of the 19th-century books in our sample.

For the entire century, the aluminon spot test showed that 91 percent of the book papers tested contained alum. Carbonate was present in only 2.8 percent of the books. The acidity, though variable for books from the first half of the 19th-century, was high for the entire century and reached an alarming level in the last three decades. Rosin began to appear in the middle of the century, and was found in about 70 percent of the books tested from the latter half of the century. Groundwood was not found in the papers from the first half of the century, but 20 percent of the books from the latter half contained groundwood.

The effect of all these factors on fold and tear resistances is clearly apparent from Appendix B, tables 4 and 6. Despite their relatively recent vintage, these papers had the lowest fold and tear resistances found in papers of any century to that date, and paper made during the last three decades of the 19th-century exhibited resistances so low in some cases as to be hardly measurable; the average for this period was truly alarming. Many very short fibers also appeared during this period.

Papermaking from 1900 to 1949: Rapidly Decreasing Permanence and Durability

The appearance of the publication *Deterioration of Book Stock—Causes and Remedies*[22] was the impetus for this and many other studies and research projects on permanence and durability of book papers. Fortunately, this renewed interest led to the development of a method for manufacturing at an affordable cost permanent and durable book papers using wood cellulose fibers.[23] However, untold damage had already been done by the publication of millions of volumes on paper that was doomed to rapid deterioration.

This section of our study is based largely on the *Deterioration* publication, which contained the results of a study of 500 books published between 1900 and 1949. The strength of the papers used in these books, especially with respect to fold endurance, was so poor that a special fold testing machine designed to test very weak papers had to be employed in order to obtain meaningful data.

In an effort to understand better the reasons for the very poor condition of these papers, we have expanded the tests originally made and published on these books to give more chemical and physical data. These data are given in Appendix E, tables 32-36. A list of the 500 test books, giving book numbers which are keyed to those in the tables, is included in Appendix D. These were nonfiction books printed and published in the United States between 1900 and 1949.

There is little or no evidence that basic changes in the papermaking process took place from 1900 to 1949. Known technology was refined and extended in terms of size and speed of machines, cost-saving improvements in pulp processing, and the like. One little-noticed observation that if capitalized upon, might have improved the useful life of paper was made by Sutermeister,[24] who found in 1901 that a promising paper could be made by incorporation of calcium carbonate, a by-product of the soda pulping process.[20] If this discovery had been followed up, an alkaline paper that would have had a longer life expectancy might have been produced much earlier, especially if a nonacid size had been invented and if there had been pressure from keepers of collections to manufacture a permanent and durable

paper. However, nothing of the sort was accomplished on a commercial basis until 1960 when Barrow developed a permanent and durable paper, made possible to an important degree by the incorporation of calcium carbonate as a mildly alkaline buffer, and by the availability of a synthetic nonacid size.

Thus the practices of the late 19th-century were in large measure continued and expanded during the 20th. High production rates and low cost were the targets. The result was that, even though these papers were the newest in our study, they were the weakest papers in terms of fold resistance of any we tested. The tear resistance was low. Acidity was high. Alum was found in 98.7 percent of the papers, and 74 percent contained rosin. From these results, we may deduce with some assurance that alum alone was used in about 24.7 percent of the papers to improve machine performance.

Carbonate was practically ignored during this fifty-year period. Only 2 percent of the papers were found to contain carbonate. Groundwood was found in 27 percent of these papers, another indication that the fibers used to make the papers were not always of the best quality.

Thus, during the first half of this century, little or no attention was paid to the production of permanent and durable book papers. Librarians and archivists are now living with the consequences. The cost of restoring or copying these books is incalculable. Moreover, the problem of durability and permance in new books is not yet solved. In spite of its availability, permanent and durable paper is still not widely accepted and used by printers and publishers.

Summary of Technical Findings, 1507 to 1949

The principal findings of our study of 1,470 books published between 1507 and 1949 can be summarized as follows:

1. Until about the middle of the 17th-century, papermaking was a tedious hand operation, more an art than a science. Certain operations were evolutionary and poorly understood, and were handed down by word of mouth rather than by written description. The relatively high quality of some of the older papers covered by the present study, beginning with 1507, was the result of careful hand operation, of the use of suitable fibers, and, in some cases, of accident (e.g., the calcium carbonate content).
2. The pressure for greater production of paper naturally led to a reduction in hand labor, introduction of machinery, use of new types of fibers, and addition of chemicals. Because of a lack of technical knowledge, the after-effects of these changes were unknown. At the time of manufacture, the paper seemed acceptable. This period marked the beginning of increasingly severe problems for permanence and durability. The principal changes which in fact contributed to the deterioration of paper quality were:
    a. The invention of the "improved" stampers and the Hollander beater, which we have shown can, if not used judiciously, produce shorter and otherwise modified fibers and, therefore, weaker paper.
    b. The use of alum (as part of the sizing, or for other reasons), which decreases the pH (increases the acidity), leading to a more rapid breakdown of the cellulose. Figures 1 and 4 show the gradual increase in the use of alum and the corresponding decrease in pH.
    c. Modifications in production processes that resulted in a decrease in the calcium carbonate content (which had counteracted acidity).
    d. The use of chlorine in bleaching colored rags and other paper-making fibers. When chlorine was originally used, its ability to degrade cellulose under certain processing conditions was not understood well enough to be controlled.[11] An excess of hypochlorite was often left in the paper as a result of over-treatment or failure to wash out the excess. In these cases, the paper was weakened.

e. The use of unpurified groundwood as a source of paper fibers. Raw groundwood as first used in papermaking was made by grinding wet wood against a revolving stone. The fibers so made were not only nonuniform in terms of length, diameter, and individuality of fibers, but also were comprised of roughly equal amounts of cellulose, the desired material, and lignin and other impurities, which resulted in a colored, weak, and rapidly deteriorating paper. It was later found that chemical purification of the wood resulted in a product containing 70 percent to 90 percent cellulose. This product is now referred to as chemical wood pulp and is the most common source of fibers for making permanent/durable paper.

Developments since 1949

The Development of Permanent/Durable Paper. Employing the results of his study of 1900-1949 book papers[22] and his knowledge of the causes of the increasingly poor conditions of book papers, W. J. Barrow began work on the development of a permanent/durable paper. The principles on which he based his efforts were:

1. Acidity is probably the principal cause of rapid deterioration of paper and in the past resulted primarily from the use of alum. Therefore, alum must not be used, and the paper must be alkaline.
2. Since wood is the cheapest and most plentiful source of cellulose fibers, it is the preferred fibrous raw material. However, since noncellulose wood components contribute importantly to poor paper permanence and durability, highly purified wood fibers would be required.
3. Short fibers contribute to the weakness of paper. Therefore, only long and strong fibers should be used.
4. A sizing material compatible with an alkaline furnish would be required.

In 1960, Barrow announced the commercial production of a slightly alkaline permanent/durable paper from wood fibers with an expected useful life of about 300 years.[23] He described the method of production and the components of the sheet, and gave tentative specifications. Several paper manufacturers are now making paper of this general type.

The materials from which this first commercial run was made were:

1. Approximately equal amounts of long, well-purified sulfate, sulfite, and soda wood fibers
2. Fillers, including small amounts of calcium carbonate, to provide an alkaline paper that would be resistant to the possible subsequent intrusion of acidic materials
3. An alkaline-compatible sizing material, Aquapel
4. A surface size of starch

Materials having these qualifications are the basis of present-day permanent/durable papers.

In order to predict the expected useful life of these papers,

methods of accelerated aging were adopted and subsequently refined.[4,7] These methods permit a projection of the useful life of paper at room temperature from tests carried out on paper which had been stored at elevated temperature and closely controlled humidity over a period varying from three days to a few weeks.

The significance of the development of commercially feasible permanent durable paper would be difficult to over-estimate. Since 1960, it has been possible to make, at a high rate of production and affordable cost, book papers equivalent in permanence and durability to those made laboriously in the 16th century. The trend toward increasingly poor paper quality shown in this study can now be reversed. Although librarians and archivists still face the monumental task of restoring or copying the tremendous number of books which are deteriorating or have deteriorated beyond use, they can take solace in knowing that there is now a way to avoid this problem in the future.

Restoration of Weak Papers. This study showed that marked deterioration in the quality of book paper began to appear about 1750. All books published since then are suspect, and many of them may be beyond use even now.

For those books printed on acidic papers that are still strong enough for use, but whose further deterioration should be retarded, Barrow described in 1959 a method of deacidification.[22] For those papers which are quite weak and not alkaline, lamination after deacidification is a solution.[5] Descriptions of other methods of deacidification have been published, and the search for new methods of deacidification and/or strengthening, including deacidification of whole books, has continued from 1959 to the present date.[10,11]

Deacidification can halt further paper deterioration resulting from acid by neutralizing the acidic components in the paper and depositing a reserve alkalinity to combat the possible intrusion of acidic materials in the future. But by itself, deacidification can not insure that the effect of other undesirable ingredients of the paper, such as high groundwood content or initially weak fibers, will be stopped. Permanence, safety, and cost are factors that must be considered in deciding which of the deacidification techniques is optimum in a given situation.

Future Needs

Further research on various aspects of paper restoration and on methods of producing permanent/durable papers is needed.

In terms of deacidifying, strengthening, and giving permanence and durability to weak papers in books, the ideal would be treatment of whole books to achieve these objectives at a reasonable cost without deleterious side effects. As a first step in this direction, cheap and reliable methods of gaseous deacidification of large numbers of whole books are currently under study in this laboratory and elsewhere. One such method is at the stage of commercial evaluation.[10]

In their efforts to encourage the use of permanent/durable paper, record custodians need a set of specifications, standards, and test methods to assure that in the future, the books and archives under their care will be printed on truly long-lasting paper. Under the sponsorship of the Council on Library Resources and the Library of Congress, this laboratory has undertaken a project to update Barrow's tentative specifications, established in 1960, for uncoated permanent/durable paper.[21] Papers from a number of manufacturers have been tested. This work indicates the requirements for a truly permanent and durable paper, and the degree to which its manufacture and use are feasible. The publication of these specifications will provide a basis on which to accept or reject papers used by publishers.

Another desirable research project is an evaluation of coated book papers, and of those containing various amounts and types of groundwood. This project should indicate whether certain paper coatings, improved methods of purifying groundwood, or the incorporation of certain additives into groundwood-containing papers can overcome the normally harmful effects of groundwood and make these papers more permanent and durable.

Cost will, of course, be a factor. But if the cost of a treatment that will make groundwood acceptable in terms of durability is not greater than the saving from the use of groundwood, a development of benefit to all parties would result.

Research on refining the correlation between accelerated and natural ageing is being, and has been, conducted in a number of laboratories. This laboratory is engaged in a long-range study of the effect of various elevated temperatures and humidities on a

number of types of book paper. A long-range study to compare natural with artificial aging is needed. It is our hope that funding can be found for the establishment of a paper archive, in which several types of book paper would be stored for long periods of time under constant and controlled conditions of normal temperature and relative humidity. The changes in physical and chemical properties could be determined periodically, say every five or ten years, and compared with those in papers artificially aged by elevated temperature and at various levels of humidity.

Summary

This study has revealed the deterioration of the permanence and durability of book papers produced during the period from 1507 to 1949, and has traced at least most of the reasons for this deterioration. It has been noted that truly permanent and durable book paper can now be made commercially at an affordable cost. Available methods and future needs for the restoration of weak papers have been described.

The cost of restoring seriously deteriorated books with very weak paper is extremely high. There is hope for relatively inexpensive deacidification of acid papers in books by use of treatments such as the gaseous morpholine deacidification process developed by this laboratory.[10] This process treating whole books can not only neutralize the acid in papers strong enough to handle, but can leave an alkaline residue which will counteract the possible subsequent intrusion of other acidic materials. There is still need for a gaseous treatment of whole books which will also strengthen very weak papers.

Failure to print books of lasting value on permanent/durable papers is folly. In the future, the cost of maintaining valuable records in usable condition will increase at a rapid rate if permanent/durable paper is not used. It is hoped that publication of this study will provide inducement to solve these problems.

In an unpublished report, Mr. George M. Cunha, Director/Conservator of the New England Document Conservation Center, has illustrated the seriousness of the problem of deterioration of library and archival materials. Mr. Cunha estimates that in an average collection, more damage is taking place in a five year span between 1970-1975 than occurred in the entire century 1700-1800. Moreover, he estimates that the rate of increase in damage with time is growing at a very rapid pace, assuming that important steps are not taken soon to arrest the decay of the records of our heritage.

This estimate should be taken very seriously by keepers of collections. If appropriate action is not taken, it is apparent that the problem of deterioration could become not only very expensive or even almost unaffordable, but that priceless documents will be lost to posterity.

While the problems of deterioration in paper made from about 1750 onward have been, or can be, solved or alleviated, the

importance of lasting records remains as John Murray expressed it in 1829.[13]

> It would certainly be difficult to point out a question of greater importance than that connected with the decay of manuscripts, and especially of valuable records . . . Time past and time present, as bearing on the relations and conditions of futurity, in all the recorded value of a world's legend—its religion and its legislative acts—its judicature and civil polity, in all their precedents—the arts—literature and science, which add lustre and renown to a nation's annals—all that exalts the human being above the brutes that perish, or the savage that roams through the desert, it is here invaded and threatened with subversion and destruction.

The list of books tested for each century (Appendix D), as well as the detailed test results for each book (Appendix E, tables 1-36), will be found on microfiche in the pocket inside the back cover of this publication. Each book tested is numbered in both the book list and the tables for cross reference.

Tables 1-5 and figures 1-5 (Appendixes B and C, respectively) were taken from Tables 1-36, and summarize the data obtained in this study.

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Appendix A:

Details of Physical and Chemical Tests

1. Folding Endurance

The outer edges of each sheet used were trimmed 1/8-1/4 in. before sampling. Three sets of 10 test strips each were cut from each book. One set was cut from the top and bottom margins; 1 set was cut from the vertical outer margins; and 1 set was cut horizontally from the printed portion of the sheets. They were designated CP, WP, and CP respectively, to indicate whether the fold line during testing would be perpendicular to the lines of print (cross printing) or parallel to them (with printing).

The 15-mm. sample strips were tested on the Massachusetts Institute of Technology Folding Endurance Tester (TAPPI, T-511), which subjected the sample to 0.5 kilogram of tension while it was being folded back and forth through 250° arcs until it broke.[16] One test was made on each strip, and the mean result is reported for each set of 10 strips. In the data tables, books are listed in order of increasing strength as measured by the average number of cross-printing folds an uninked strip would withstand before breaking.

2. Tear Resistance

After the edges of the sample sheets were trimmed, standard 63-mm. tear test specimens were cut from them without regard to the presence or absence of printing.

Samples from each book were tested in both cross-printing and with-printing directions to determine their resistance to tear due to shear stresses such as those imposed during page turning. The Elmendorf Tear Resistance Tester (TAPPI, T-414), which measures the force required to tear 43 mm. through a stack of samples that has been previously notched, was employed.[18] Each tear resistance value reported is the average from five 8-ply tests and gives, in grams, the force which would be required to tear through 16 plies of that paper.

3. Alum, Rosin, and Groundwood Content

The spot tests described by Barrow[2] were used to detect the presence of these constituents:

Alum. Aluminon solution (1 gm. per liter of distilled water) was applied thinly to the paper surface. Where no alum was present the spot remained a faint pink, but the presence of the aluminum ion produced a deep pink color. A purplish reaction indicated the presence of iron.

Rosin. A drop of nearly saturated sugar solution was applied to an uninked paper surface and excess liquid blotted off. Onto this spot 96.6% sulfuric acid was spread with a medicine dropper. If rosin was present the spot quickly developed a pink raspberry color. Where there was no rosin, the spot remained colorless or turned a brownish color. In a few cases, where a substantial amount of groundwood was present, the paper blackened, obscuring the possible development of a pink raspberry color. In these cases, the results of the rosin test are indicated by a question mark in the rosin column in Appendix E.

Groundwood. The test reagent was 1 gram of phloroglucinol in 50 ml. of methanol and 50 ml. concentrated hydrochloric acid. The reagent was applied thinly to an uninked paper surface with a dropper or glass rod and, if groundwood was present, the treated paper immediately turned a deep purplish red. The spot remained colorless in the absence of groundwood. (The solution darkens with age and produces different color reactions; though refrigeration will greatly prolong its life, fresh solution should be made periodically.)

4. Metal Carbonates

Test specimens were first immersed in distilled water to expel trapped air and then immersed in about 4% aqueous hydrochloric acid solution. If an evolution of bubbles (carbon dioxide from the carbonate) from the surface of the paper was observed upon immersion in the acid solution, the test was considered positive.

5. pH

The pH of the papers was determined by the Technical Association of the Pulp and Paper Industry (TAPPI) cold extraction method (T-509), using a Beckman Zeromatic pH Meter.[11] The sample was cut into squares of 1/8 by 1/8 in. or less, and 1 gram of these was soaked in distilled water for a minimum of 1 hour. The pH of the water extract was then determined directly with the calibrated meter.

6. Fiber Analysis

The uninked area from at least 2 pages in each book was used for fiber analyses. The paper, after removal from the book was torn into smaller pieces and put into a blender with enough of a 1% solution of sodium hydroxide solution to cover the blades of the blender. The blender was operated at a low speed until the paper became pulpy.

The fibers were then poured into a small, fine-mesh strainer, and tap water was run over them to wash out the sodium hydroxide solution. The fibers were squeezed until the excess water was removed.

While the fibers were still in the strainer, they were wetted with a 2% hydrochloric acid solution and allowed to stand for approximately 2 minutes. The fibers were rinsed thoroughly with tap water, and the excess water was squeezed out. The fibers were then dispersed in clean water.

An eyedropper, with a large opening at the bottom to assure the capture of large as well as small fibers, was used to sample the fibers. One or 2 drops of the slurry was placed on a microscope slide. The water was then allowed to evaporate from the fibers on the slide.

After the water evaporated, the proper stain was applied to the fibers. "Herzberg" and "C" stains were used for identifying chemical wood, groundwood, and rag by color reactions. Stain charts are available for this purpose. (John H. Groff, Color Atlas for Fiber Identification. Appleton, Wisconsin: Institute of Paper Chemistry. 1940.)

A binocular microscope offering magnifications of 60, 100, and 430 was used. Polarized light was used to distinguish the characteristics of cotton and linen fiber. Methyl salicylate was used as the mounting medium.[3] The lengths of 100 fibers were measured with a scale incorporated in the microscope eyepiece.

Abbreviations Used in the Tables

Area of Paper
I—Inked (printed area)
U—Uninked (unprinted area)

Direction of Test
CP—Cross printing, i.e., line of test at right angles to the lines of printing, parallel to the spine.
WP—With printing, i.e., line of test parallel to the lines of printing, perpendicular to the spine.

Fiber
L—Length in millimeters
CW—Chemical wood
GW—Groundwood
HW—Hardwood
R—Rag
S—Straw
SW—Softwood

Folding Test
MIT—Massachusetts Institute of Technology Fold Endurance Tester

Tear Test
Elmendorf—Elmendorf Tear Tester

Letters following specimen numbers
A—American publication
B—British publication
E—Continental publication

Additives
R—Rosin
C—Carbonate
A—Aluminum
G—Groundwood
P—Positive, i.e., additive present
N—Negative, i.e., additive not present

Basis Weight
BW—Weight in pounds of a ream of 500 sheets, 25 x 38 in. each

Appendix 8: Summary Tables of Data on Book Papers, 1507 to 1949

Table 1

[illegible]

[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]
[illegible]	[illegible]

Table 2

[illegible]

[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	Alum		Carbonate		Rosin		Ground-wood	
					Sngl.	Pos.	Sngl.	Pos.	Sngl.	Pos.	Sngl.	Pos.
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	4	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	4	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	3	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	4	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	4	[illegible]	[illegible]	[illegible]	-	-	-	-
	[illegible]				[illegible]	[illegible]	[illegible]	[illegible]				
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	1	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	2	[illegible]	9	1	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	4	[illegible]	8	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	9	1	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	8	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	44	1	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	44	1	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	-	-	-	-
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		-	-	-
	[illegible]				[illegible]	[illegible]	[illegible]	[illegible]				

Table 2 - Continued

Median Values of Physical Data, by Decade

Decade	Total No. Books Tested	Fold (un-inked CM)	Tear (un-inked CP)	pH (un-inked)	Alum No. Neg.	Alum No. Pos.	Carbonate No. Neg.	Carbonate No. Pos.	Rosin No. Neg.	Rosin No. Pos.	Groundwood No. Neg.	Groundwood No. Pos.
1700-09	25	505	41.6	5.3	7	18	21	4	-	-	-	-
1710-19	25	97	47.6	5.1	6	19	22	3	-	-	-	-
1720-29	25	112	49.4	5.6	11	14	17	8	-	-	-	-
1730-39	25	89	45.4	5.0	4	21	22	3	-	-	-	-
1740-49	25	180	38.6	4.7	3	22	22	3	-	-	-	-
1750-59	25	[illegible]	34.0	4.6	1	24	25	0	-	-	-	-
1760-69	25	43	41.0	4.5	0	25	25	0	-	-	-	-
1770-79	25	90	38.0	4.6	2	21	25	0	-	-	-	
1780-89	25	[illegible]	37.1	4.4	1	24	25	0	-	-	-	-
1790-99	25	72	34.2	4.5	2	23	24	1	-	-	-	-
	250				37	213	228	22				
1800-09	50	18	28.7	4.6	3	47	49	1	50	0	50	0
1810-19	50	22	23.6	4.7	5	45	[illegible]	0	50	0	50	0
1820-29	50	27	30.2	5.2	8	42	49	1	50	0	50	0
1830-39	50	35	30.4	[illegible]	8	42	44	6	49	1	50	0
1840-49	50	35	36.0	[illegible]	7	43	49	1	45	5	50	0
1850-59	50	15	33.4	5.1	6	44	47	3	34	16	49	1
1860-69	50	8	22.2	4.4	6	44	48	2	29	21	47	3
1870-79	50	3	18.6	4.3	1	49	[illegible]*	0*	3	47	31	19
1880-89	50	2	19.5	[illegible]	0	50	50	0	2	48	37	13
1890-99	50	2	20.9	4.4	0	50	50	0	1	49	36	14
	500				44	456	48[illegible]	14	313	187	450	50
1900-09	100	4**	25.7	[illegible]	0	100	100	0	20	80	74	26
1910-19	100	3**	24.7	4.3	1	97	98	2	23	77	75	25
1920-29	100	4**	26.2	[illegible]	0	100	100	0	47	53	79	21
1930-39	100	9**	37.4	4.8	0*	99*	[illegible]*	2*	22*	77*	73*	26*
1940-49	[illegible]	19**	41.5	4.7	5*	96*	[illegible]*	6*	15*	84*	61*	38*
	500				6	492	488	10	127	371	362	136

*Some of these books were not available for all of the tests that were run because the supply of paper was depleted.

**Because of the weakness of the papers, a modified MIT fold tester was used. This device swings the paper strips in consecutive 90° arcs under 1 kg. tension. The number of folds would have been much smaller if the standard MIT fold tester could have been used.

Table 3

Average Median Values of Physical Data, by Century

Years	Total No. of Books Tested	Avg. Median Fold (uninked CP)	Avg. Median Tear (uninked CP)	No. of Books with pH 6.0 and above	Percent	No. of Books with pH 5.0-5.9	Percent	No. of Books with pH 4.9 and below	Percent
1500-99	45	440	43.7	37	82%	7	16%	1	2%
1600-99	175	361	42.1	56	32%	74	42%	45	26%
1700-99	250	129	40.8	38	15%	64	26%	148	59%
1800-99	500	17	26.3	79	16%	132	26%	289	58%
1900-49	500	8*	31.1	47	9%	140	28%	311	62%

*Because of the weakness of the papers, a modified MIT fold tester was used. This device swings the paper strips in consecutive 90° arcs under 1 kg. tension. The number of folds would have been much smaller if the standard MIT fold tester could have been used.

Table 4

Average Median Decade Values with Highest and Lowest Median Values for Fold, Tear, and pH, Recorded by Century, 1507-1899

		Fold			Tear			pH		
Century	No. of Books Tested	Avg. of 10 Median Decade Values	High Median Value	Low Median Value	Avg. of 10 Median Decade Values	High Median Value	Low Median Value	Avg. of 10 Median Decade Values	High Median Value	Low Median Value
1507-1599	45	440	[illegible]	72	[illegible]	63.0	36.2	6.7	8.5	5.6
1600-1699	[illegible]	361	727	144	42.1	49.6	37.2	[illegible]	6.8	5.2
1700-1799	[illegible]	129	505	43	40.8	49.4	34.0	4.8	[illegible]	4.4
1800-1899	500	17	35	2	26.3	36.0	18.6	4.9	5.8	4.5

Table 5

Average Median Decade Values with Highest and Lowest Median Values for Fold, Tear, and pH, Recorded by Half-Century, 1700-1949

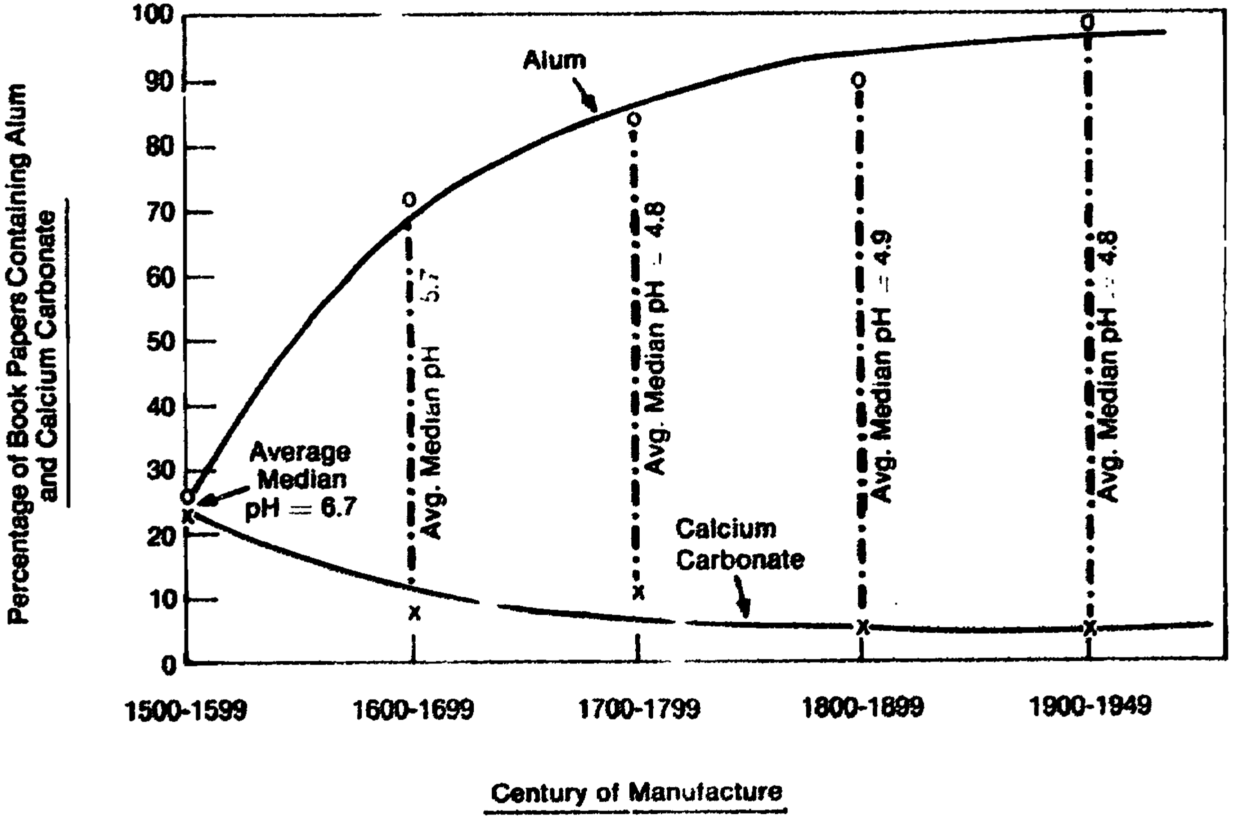
		Fold			Tear			pH		
Half-Century	No. of Books Tested	Avg. of 5 Median Decade Values	High Median Value	Low Median Value	Avg. of 5 Median Decade Values	High Median Value	Low Median Value	Avg. of 5 Median Decade Values	High Median Value	Low Median Value
1700-1749	125	197	505	89	44.5	49.4	38.6	5.1	5.6	4.7
1750-1799	125	62	90	43	37.0	41.0	34.0	4.5	4.6	4.4
1800-1849	250	27	35	18	29.7	36.0	23.6	5.1	5.8	4.6
1850-1899	250	6	15	2	22.9	33.4	18.6	4.7	5.1	4.5
1900-1949	500	8*	14*	3*	31.1	41.5	24.7	4.8	4.9	4.7

*Because of the weakness of the papers, a modified MIT fold tester was used. This device swings the paper strips in con[illegible] 90° arcs under 1 kg. tension. The number of folds would have been much smaller if the standard MIT fold tester could have been used.

Appendix C: Figures

Figure 1

Alum and Calcium Carbonate in Book Papers (1507-1949)



Fold Endurance of Book Paper (1507-1949)

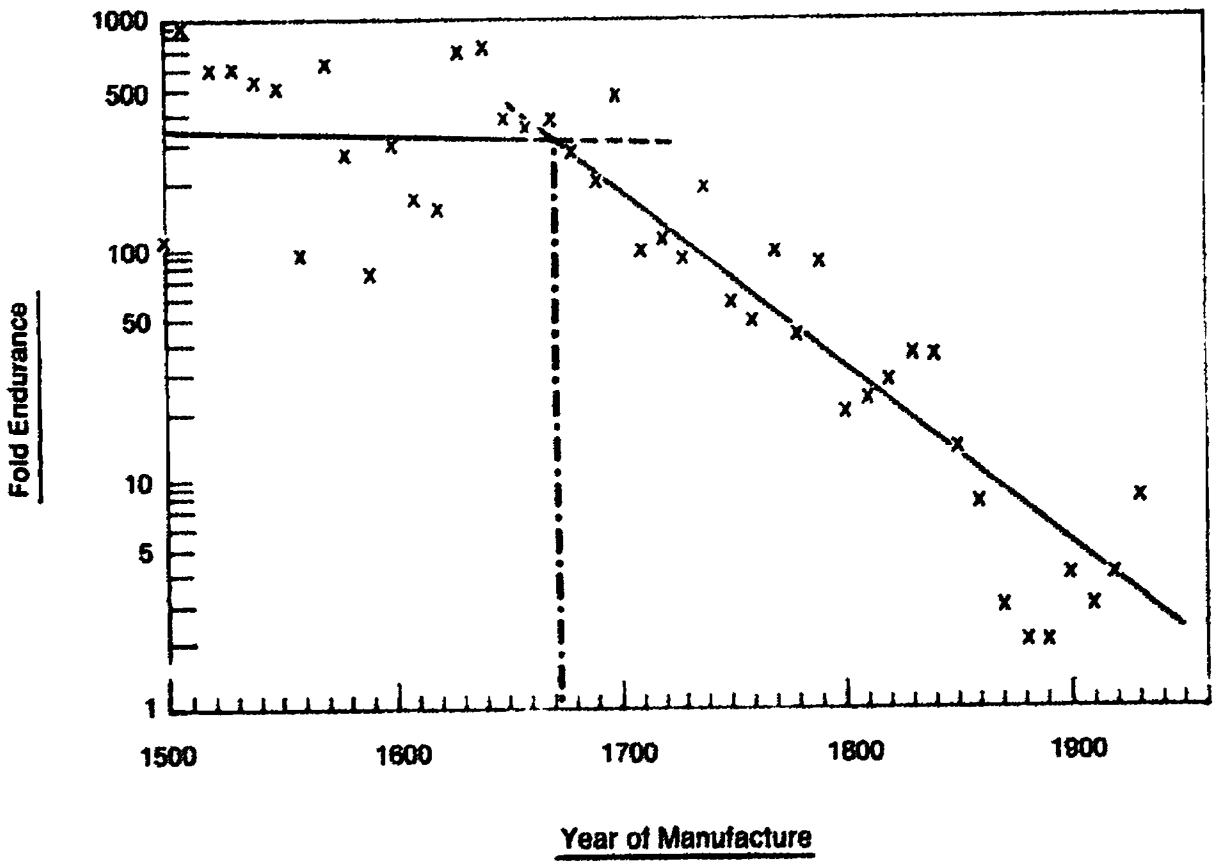
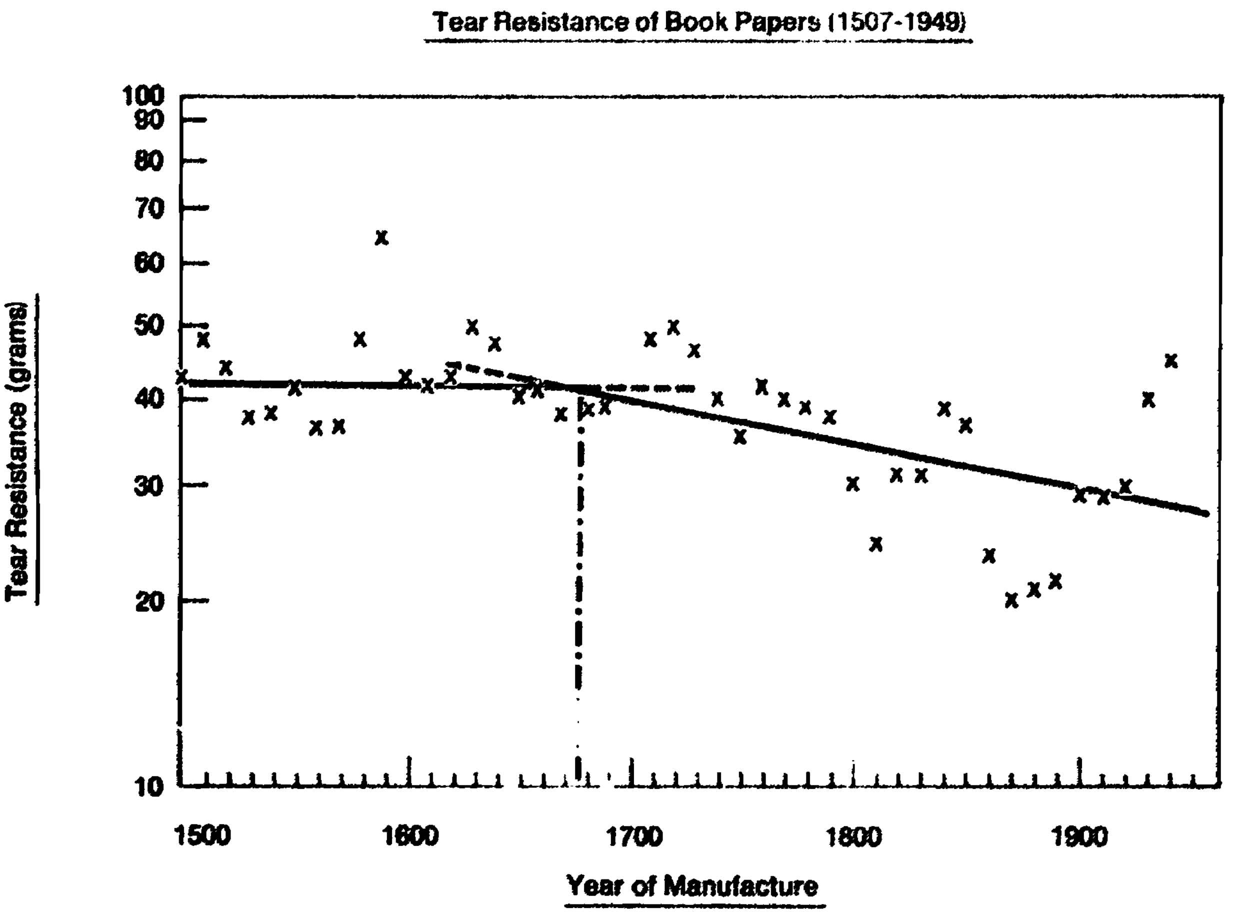


Figure 2

Figure 3



Tear Resistance of Book Papers (1507-1949)

Figure 4

pH of Book Papers (1507-1949)

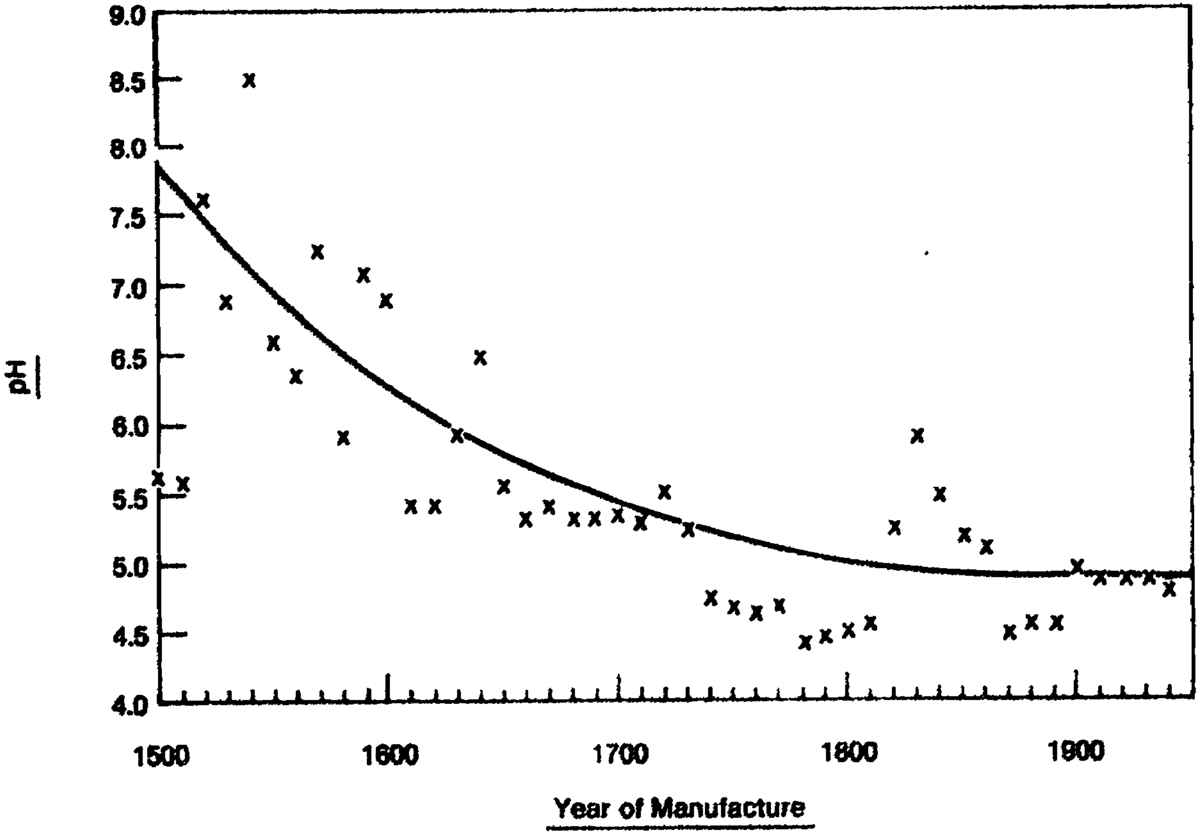
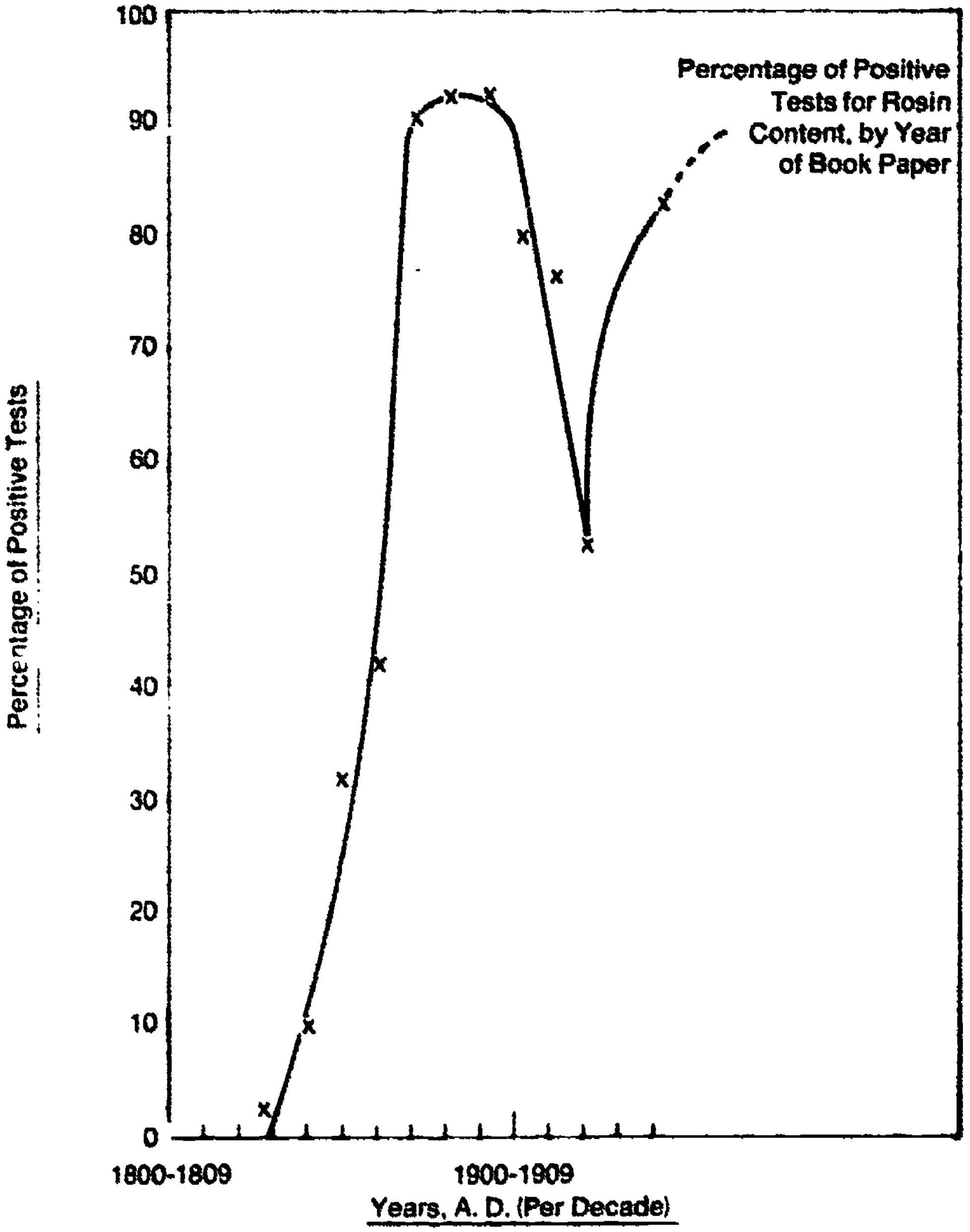


Figure 5


100
90
80
70
60
50
40
30
20
10
0
Percentage of Positive Tests
Percentage of Positive Tests for Rosin Content, by Year of Book Paper
1800-1809
1900-1909
Years, A. D. (Per Decade)